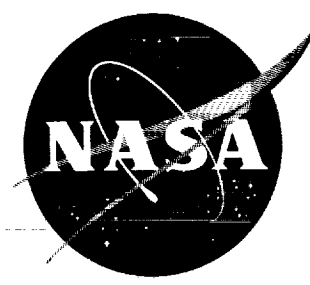


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TECHNICAL NOTE

D-1403

THERMODYNAMIC CONSISTENCY OF SOLUBILITY DATA FOR
THE HYDROGEN-HELIUM VAPOR-LIQUID SYSTEM

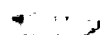
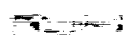
By Irving Brazinsky and Byron S. Gottfried

Lewis Research Center
Cleveland, Ohio

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TECHNICAL NOTE D-1403

THERMODYNAMIC CONSISTENCY OF SOLUBILITY DATA FOR

THE HYDROGEN-HELIUM VAPOR-LIQUID SYSTEM

By Irving Brazinsky and Byron S. Gottfried

SUMMARY

The solubilities of helium in liquid hydrogen at several temperatures and pressures have been experimentally obtained by S. R. Smith at Ohio State University. Smith was unable analytically to show thermodynamic consistency in his data. This inconsistency appears to have been produced by the use of incorrect values for the coefficients in a virial equation of state. In this report these data were evaluated for thermodynamic consistency by determination of hydrogen fugacities by two independent techniques, one of which employs recently obtained experimental values for the required virial coefficients. The solubility data are reasonably consistent, although the accuracy of the analysis is somewhat limited by simplifying assumptions. Both the original solubility data and the analysis of thermodynamic consistency are presented herein.

INTRODUCTION

Gaseous helium is currently used to pressurize liquid-hydrogen-rocket tankage systems, and concern has been expressed concerning the solubility of helium in liquid hydrogen. Reference 1 by S. R. Smith contains experimental equilibrium data for the hydrogen-helium system at three temperatures (17.4° , 20.39° , and 21.8° K) and at pressures ranging from 43 to 475 pounds per square inch absolute. These data indicate that the solubility of helium in liquid hydrogen is small.

Smith also attempted (ref. 1) to evaluate the thermodynamic consistency of his data, but his calculated values of the hydrogen gas-phase fugacities are in wide disagreement with the values of the hydrogen liquid-phase fugacities. This discrepancy between the fugacities caused some doubt about the reliability of the data.

In calculating hydrogen gas-phase fugacities, Smith employed a method requiring the use of the second interaction virial coefficient. Because experimental data for this virial coefficient were not available, the required values were calculated. Since the completion of reference 1, however, experimental values of the second interaction virial

coefficient have been obtained for the hydrogen-helium system (ref. 2) that are appreciably different from Smith's values. The thermodynamic consistency of Smith's solubility data are therefore evaluated herein by utilizing the experimentally determined values of the second virial coefficients. The important solubility data from reference 1 are also included herein.

PRESENTATION OF DATA

The temperature, total pressure, and liquid-vapor compositions were measured by Smith (ref. 1) at various pressures along three isotherms: 17.4°, 20.39°, and 21.8° K. Pressures ranged from 48 to 316 pounds per square inch absolute at 17.4° K, from 63 to 475 pounds per square inch absolute at 20.39° K, and from 43 to 386 pounds per square inch absolute at 21.8° K. The data from reference 1 are presented in table I, and pressure-composition diagrams are shown in figures 1(a), (b), and (c) for temperatures of 17.4°, 20.39°, and 21.8° K, respectively. The phase rule indicates that a two-phase binary system, which the hydrogen-helium liquid-vapor system is, contains two degrees of freedom. Thus, if the pressure and the temperature of such a system are specified, the liquid- and gas-phase compositions are uniquely determined. As an illustration, at 17.4° K and 200 pounds per square inch absolute, the liquid is 0.66 mole percent helium and the gas in equilibrium with this liquid is 94.5 mole percent helium (fig. 1(a)).

Smith found that, at each temperature, a plot of the fugacity of helium against the mole fraction of helium in the liquid was a straight line (see fig. 2). This straight line indicates that Henry's law (ref. 3), which states that the fugacity of the solute in a two-phase mixture varies linearly with the concentration of that constituent in the liquid, could be applied to the hydrogen-helium equilibrium mixture. Because the gas phase is not a perfect gas mixture, the fugacity in Henry's law is used in place of partial pressure.

ANALYSIS

The approach taken herein in analyzing the data involves a comparison between the fugacities of liquid hydrogen and of gaseous hydrogen as determined by two independent techniques. In the first method, Henry's law was substituted into the Gibbs-Duhem equation, and the resulting expression was integrated to obtain a relation among pressure, liquid-phase composition, and liquid-phase fugacity. In the second method, a virial equation of state was substituted into an exact thermodynamic expression for the fugacity of a component in a gas mixture; thus, the fugacity of hydrogen vapor was obtained as a function of vapor-phase composition. Consistent data should result in agreement between the values of the fugacities calculated from the two methods, since the liquid- and gas-phase fugacities must be equal in an equilibrium mixture.

Liquid-Phase Fugacities

The Gibbs-Duhem equation is given in reference 4 as

$$\left(\frac{\partial \ln f_1}{\partial \ln x_1} \right)_T = \left(\frac{\partial \ln f_2}{\partial \ln x_2} \right)_T + \frac{V'_m}{RT} \frac{dP}{dx_1} \quad (1)$$

This equation may be put into the more convenient form

$$x_1 \frac{df_1}{f_1} + (1 - x_1) \frac{df_2}{f_2} = \frac{V'_m}{RT} dP \quad (2)$$

It has been experimentally shown (ref. 1) that the solute obeys Henry's law (fig. 2), which is given by

$$f_1 = kx_1$$

Thus,

$$df_1 = k dx_1$$

and

$$\frac{df_1}{f_1} = \frac{dx_1}{x_1} \quad (3)$$

Substituting equation (3) into equation (2) yields

$$dx_1 + (1 - x_1) \frac{df_2}{f_2} = \frac{V'_m}{RT} dP$$

Replacing $(1 - x_1)$ by x_2 and dx_1 by $-dx_2$ gives

$$\frac{1}{f_2} \frac{df_2}{dx_2} = \frac{1}{x_2} + \frac{V'_m}{x_2 RT} \frac{dP}{dx_2}$$

or

$$-\frac{d \ln f_2}{dx_2} = \frac{1}{x_2} + \frac{V'_m}{x_2 RT} \frac{dP}{dx_2}$$

Since dilute solutions are involved, x_2 is approximately equal to 1; therefore,

$$-\frac{d \ln f_2}{dx_2} = \frac{1}{x_2} + \frac{V'_m}{RT} \frac{dP}{dx_2} \quad (4)$$

Smith used equation (4) to determine $\ln f_2$ as a function of x_2 and dP/dx_2 . His method of obtaining fugacities involved the determination of dP/dx_2 from the slopes obtained from a smoothed curve representing experimental equilibrium data points.

Formal integration of equation (4) is desirable so that the analysis will be independent of an estimated slope. This integration, with the assumption that V'_m (the molal volume of the liquid) is constant, is

$$\int_{f_2^o}^{f_2} \frac{df_2}{f_2} = \int_1^{x_2} \frac{dx_2}{x_2} + \frac{V'_m}{RT} \int_{p_2^o}^P dP$$

or

$$\ln \frac{f_2}{f_2^o} = \ln x_2 + \frac{V'_m}{RT} (P - p_2^o)$$

or

$$f_2 = x_2 f_2^o \exp \left[\frac{V'_m}{RT} (P - p_2^o) \right] \quad (5)$$

Equation (5) is used herein to calculate values of f_2 corresponding to measured values of x_2 and P at a given temperature.

Gas-Phase Fugacities

Smith used the Lewis and Randall rule, $f_1 = f_{g,1} y_1$, to calculate the fugacities of helium (ref. 3). The fugacities of pure gaseous helium ($f_{g,1}$) were evaluated at the temperature and the total pressure of the system. According to Smith, the Lewis and Randall rule accurately predicts the gas-phase fugacity of helium because the temperature

of the system is always above the critical temperature of helium. Thus, values of $f_{g,1}$ do not have to be obtained by extrapolating into the liquid region.

The gas-phase fugacities of hydrogen could not be calculated accurately by the Lewis and Randall rule because at every point at which equilibrium measurements were made, the temperature and the total pressure of the system were such that pure hydrogen was a subcooled liquid. The value of $f_{g,2}$ would therefore be obtained by an extrapolation of the gaseous-hydrogen fugacity curve into the liquid region.

Smith calculated the gas-phase fugacities of hydrogen from the following expression:

$$\ln f_{\text{mix}} = x_1 \ln \frac{f_1}{x_1} + x_2 \ln \frac{f_2}{x_2}$$

where f_1 (the fugacity of helium) is known from the Lewis and Randall rule, and f_{mix} is the fugacity of the gas mixture. Smith, however, does not state how he obtained f_{mix} except to mention that the calculation of f_{mix} is dependent on a knowledge of the second virial coefficients. The values of the hydrogen gas-phase fugacities obtained by Smith in this manner are listed in table I. Because the logical development of Smith's method for determining hydrogen gas-phase fugacities is not clear and the Lewis and Randall rule does not apply, an independent method is developed herein for obtaining the gas-phase fugacities of hydrogen using Smith's solubility data.

An exact thermodynamic expression for the fugacity of a component in a gas mixture is given in reference 5 as

$$RT \ln f_2 = \int_0^P \left(\frac{\partial V}{\partial n_2} - \frac{RT}{P} \right) dP + RT \ln (y_2 P) \quad (6)$$

An expression that allows the explicit use of equation (6) to evaluate the fugacity of a component in a gas mixture is derived as follows. The derivation employs a modified form of the equation of state:

$$\frac{Pv}{RT} = 1 + \frac{B}{v} + \frac{C}{v^2} + \frac{D}{v^3} + \dots \quad (7)$$

where B , C , D , etc. are temperature-dependent virial coefficients.

The form of this equation used herein is

$$\frac{Pv}{RT} = 1 + B'P + C'P^2 + \dots \quad (8)$$

where $B' = B/RT$ and $C' = (C - B^2)/(RT)^2$ (ref. 6). As a truncated polynomial, equation (8) is a slightly less accurate form of the virial equation of state than is equation (7), but because of the subsequent mathematical manipulations, equation (8) is a more desirable form.

At the pressures employed in the investigation, terms beyond $B'P$ are assumed to be negligible because of the extremely small values of the third, fourth, etc. virial coefficients. Equation (8) reduces to

$$\frac{Pv}{RT} = 1 + B'P = 1 + \frac{BP}{RT} \quad (9)$$

For binary mixtures,

$$PV = (n_1 + n_2)(RT + B_m P) \quad (10)$$

and

$$v = \left(\frac{n_1 + n_2}{P} \right) (RT + B_m P)$$

where

$$B_m = y_1^2 B_1 + 2y_1 y_2 B_{12} + y_2^2 B_2$$

Thus

$$v = \left(\frac{n_1 + n_2}{P} \right) \left[RT + P(y_1^2 B_1 + 2y_1 y_2 B_{12} + y_2^2 B_2) \right] \quad (11)$$

The partial molal volume of component 2 (hydrogen) is obtained by differentiation of equation (11) with respect to n_2 , with n_1 , T , and P constant:

$$\frac{\partial v}{\partial n_2} = \frac{RT}{P} + B_m + (n_1 + n_2) \frac{\partial B_m}{\partial n_2}$$

By the chain rule for partial differentiation,

$$\begin{aligned}\frac{\partial B_m}{\partial n_2} &= \frac{\partial B_m}{\partial y_1} \frac{\partial y_1}{\partial n_2} + \frac{\partial B_m}{\partial y_2} \frac{\partial y_2}{\partial n_2} \\ &= \left(\frac{\partial B_m}{\partial y_2} - \frac{\partial B_m}{\partial y_1} \right) \left(\frac{y_1}{n_1 + n_2} \right)\end{aligned}$$

From evaluation of the derivatives, it is easily seen that

$$(n_1 + n_2) \frac{\partial B_m}{\partial n_2} = 2y_1(y_1B_{12} + y_2B_2 - y_1B_1 - y_2B_{12})$$

Adding B_m to this equation and rearranging results in

$$B_m + (n_1 + n_2) \frac{\partial B_m}{\partial n_2} = 2y_2B_2 + 2y_1B_{12} - B_m$$

Therefore,

$$\frac{\partial V}{\partial n_2} = \frac{RT}{P} + 2y_2B_2 + 2y_1B_{12} - B_m \quad (12)$$

Substituting equation (12) into equation (6) and integrating gives

$$\ln f_2 = \ln (y_2P) + \frac{P}{RT} (2y_2B_2 + 2y_1B_{12} - B_m)$$

or

$$f_2 = y_2P \exp \left[\frac{P}{RT} (2y_2B_2 + 2y_1B_{12} - B_m) \right] \quad (13)$$

Equation (13) can be used to calculate values of f_2 corresponding to y_2 and P at a given temperature, providing the required second virial coefficient data are available.

Smith obtained values of gas-phase hydrogen fugacities from calculated values of gas-phase helium fugacities and calculated values for

the second interaction virial coefficient B_{12} . The values used by Smith for the second virial coefficients are:

Temperature, T , $^{\circ}\text{K}$	Second virial coefficient, cc/g-mole		
	Hydrogen, B_2	Helium, B_1	Interaction, B_{12}
17.4	-200.5	-4.99	-77.5
20.39	-147.5	-2.08	-58
21.8	-131	-13.38	-50.3

Recently, however, experimental values of second virial coefficients for hydrogen-helium gas mixtures were published by Varekamp and Beenakker (ref. 2). The values of the virial coefficients obtained from reference 2 are:

Temperature, T , $^{\circ}\text{K}$	Second virial coefficient, cc/g-mole		
	Hydrogen, B_2	Helium, B_1	Interaction, B_{12}
17.4	-190	-7.0	-23.9
20.39	-151.3	-3.7	-16.2
21.8	-141	-2.5	-13.6

The values of B_2 and B_1 used by Smith are approximately equal to the values listed in this table, except for the value of B_1 at 21.8°K . The values of B_{12} , however, differ greatly from the values obtained in reference 2.

The gas-phase fugacities of hydrogen presented herein were calculated from equation (13) by utilizing the second virial coefficients from reference 2.

DISCUSSION OF RESULTS

The equilibrium data of Smith would be considered thermodynamically consistent if the gas-phase fugacities determined from the measured gas-phase compositions and equation (13) agreed with the liquid-phase fugacities that were obtained from the experimentally determined liquid-phase compositions and equation (5). The values obtained by using these

two methods agree to within about 17 percent (based upon the liquid-phase fugacities) for the 17.4° K isotherm, 21 percent for the 20.39° K isotherm, and about 24 percent for the 21.8° K isotherm when the second virial coefficients from reference 2 are used. The discrepancies among these values are believed to be due to Smith's equilibrium data; however, they are not unreasonably large. The calculated values for the gas-phase and liquid-phase fugacities are listed with their respective equilibrium pressures in table I for the three temperatures and are shown graphically in figure 3. Smith's calculated fugacities are also included in figure 3.

From figure 3, the agreement between the calculated fugacities is clearly better for the 17.4° and 20.39° isotherms than for the 21.8° isotherm. Even a cursory glance at figure 3, however, will reveal that, in all three cases, the agreement between gas- and liquid-phase fugacities obtained by using the values of the gas-phase second virial coefficients from reference 2 and the method developed herein is far superior to the agreement that Smith obtained.

The method of checking the thermodynamic consistency of the data presented herein has two limitations:

(1) Generally, the fugacity of a component in a gas mixture is difficult to calculate when the temperature of the mixture is below the critical temperature of that component. A more accurate representation of state data than are available herein for the hydrogen-helium mixture may enable more accurate calculations of the hydrogen-gas fugacities.

(2) Another possible source of uncertainty is the set of assumptions involved in the derivation of equation (5) for the liquid-phase fugacity. To obtain equation (5), V_m' is assumed constant and hence invariant with pressure and composition at constant temperature. The liquid is essentially hydrogen, and the molal volume of liquid hydrogen does vary to a slight degree through the range of pressures investigated herein. In addition, x_2 was considered constant, although this quantity also varied slightly over the pressure range considered. It was further assumed that Henry's law applies for the helium fugacities in the liquid phase over the entire range of pressures.

The errors introduced into the analysis by the second limitation are believed to be small compared with the errors introduced by the first limitation.

CONCLUDING REMARKS

In view of the previous analysis, it may be concluded that the hydrogen-helium vapor-liquid solubility data presented by Smith (ref. 1)

are fairly reliable. The apparent lack of thermodynamic consistency which Smith originally reported seems to be due to the use of calculated values for the second virial interaction coefficient that were inaccurate. These values were used to determine the gas-phase fugacities of hydrogen from experimental solubility data. The use of later, experimentally determined values for the second virial interaction coefficient in the equations derived herein resulted in a reasonably accurate check on the reliability of the solubility data.

Lewis Research Center

National Aeronautics and Space Administration
Cleveland, Ohio, April 20, 1962

APPENDIX - SYMBOLS

B	second virial coefficient, cc/g-mole
B_m	second virial coefficient of mixture, cc/g-mole
B_{12}	second interaction virial coefficient, cc/g-mole
f	fugacity, atm
f_2^0	fugacity of pure saturated liquid hydrogen evaluated at temperature and vapor pressure of liquid, atm
f_g	fugacity of pure gas, evaluated at the temperature and total pressure of the system, atm
f_{mix}	fugacity of gas mixture, atm
k	Henry's law constant, atm
n	number of moles of component in gas phase
P	total pressure of system, atm
P_2^0	vapor pressure of liquid hydrogen, atm
R	universal gas constant, (atm)(cc)/(g-mole)(°K)
T	absolute temperature, (°K)
V	total volume of gas, cc
V'_m	molar volume of liquid phase, cc/g-mole of mixture
v	specific volume of gas, cc/g-mole
x	mole fraction in liquid
y	mole fraction in vapor

Subscripts:

1	solute, helium
2	solvent, hydrogen

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TABLE I. - HYDROGEN-HELIUM EQUILIBRIUM DATA

(a) Temperature, 17.4° K; vapor pressure of liquid hydrogen, 0.362 atmosphere; fugacity of liquid hydrogen, 0.345 atmosphere

Pressure, lb/sq in. abs	Helium in liquid, mole percent (a)	Helium in vapor, mole percent (a)	Fugacity of helium, atm (a)	Hydrogen in liquid, mole percent	Gas-phase fugacity of hydrogen, atm (a)	Liquid-phase fugacity of hydrogen, atm (b)	Gas-phase fugacity of hydrogen, atm (c)
48	0.20	83.5	2.70	100	0.371	0.367	0.345
83	.22	89.7	4.97	99.8	.314	.384	.443
96	.27	91.0	5.80	99.78	.289	.388	.440
141	.45	93.5	8.70	99.73	.218	.410	.432
207	.74	94.6	12.70	99.55	.170	.446	.417
316	1.08	95.1	19.1	99.26	.0965	.513	.435
				98.92			.470

^aRef. 1.

^bEq. (5).

^cCalculated from eq. (13) with virial coefficients of ref. 2.

TABLE I. - Continued. HYDROGEN-HELIUM EQUILIBRIUM DATA

(b) Temperature, 20.39° K; vapor pressure of liquid hydrogen, 1.00 atmosphere; fugacity of liquid hydrogen, 0.915 atmosphere

Pressure, lb/sq in. abs	Helium in liquid, mole percent (a)	Helium in vapor, mole percent (a)	Fugacity of helium, atm (a)	Hydrogen in liquid, mole percent	Gas-phase fugacity of hydrogen, atm (a)	Liquid-phase fugacity of hydrogen, atm (b)	Gas-phase fugacity of hydrogen, atm (c)
63	0.59	73.7	3.14	100	0.85	0.96	0.914
102	.87	81.0	5.56	99.13	.805		.905
177	1.53	87.3	10.4	98.47	.635		
197	1.63	88.2	11.7	98.37	.595	1.110	1.01
222	2.04	88.9	13.3	97.96	.542		
256	2.16	89.6	15.4	97.84	.498		
257	2.18	89.6	15.5	97.82	.497	1.185	1.05
291	2.47	89.9	17.5	97.53	.457		
306	2.48	90.0	18.4	97.52	.436		
311	2.55	90.0	18.8	97.45	.431	1.255	1.085
317	2.66	90.0	19.1	97.34	.429		
344	2.82	90.0	20.8	97.18	.398		
350	2.77	90.0	21.1	97.23	.394	1.305	1.110
358	2.77	90.0	21.6	97.23	.384		
365	2.95	90.0	22.0	97.05	.376		
375	2.94	90.0	22.6	97.06	.366	1.34	1.14
396	3.01	90.0	23.9	96.99	.344		
406	3.06	90.1	24.6	96.94	.328		
475	3.44	90.1	28.6	96.56	.178	1.510	1.19

^aRef. 1.

^bEq. (5).

^cCalculated from eq. (13) with virial coefficients of ref. 2.

TABLE I. - Concluded. HYDROGEN-HELIUM EQUILIBRIUM DATA

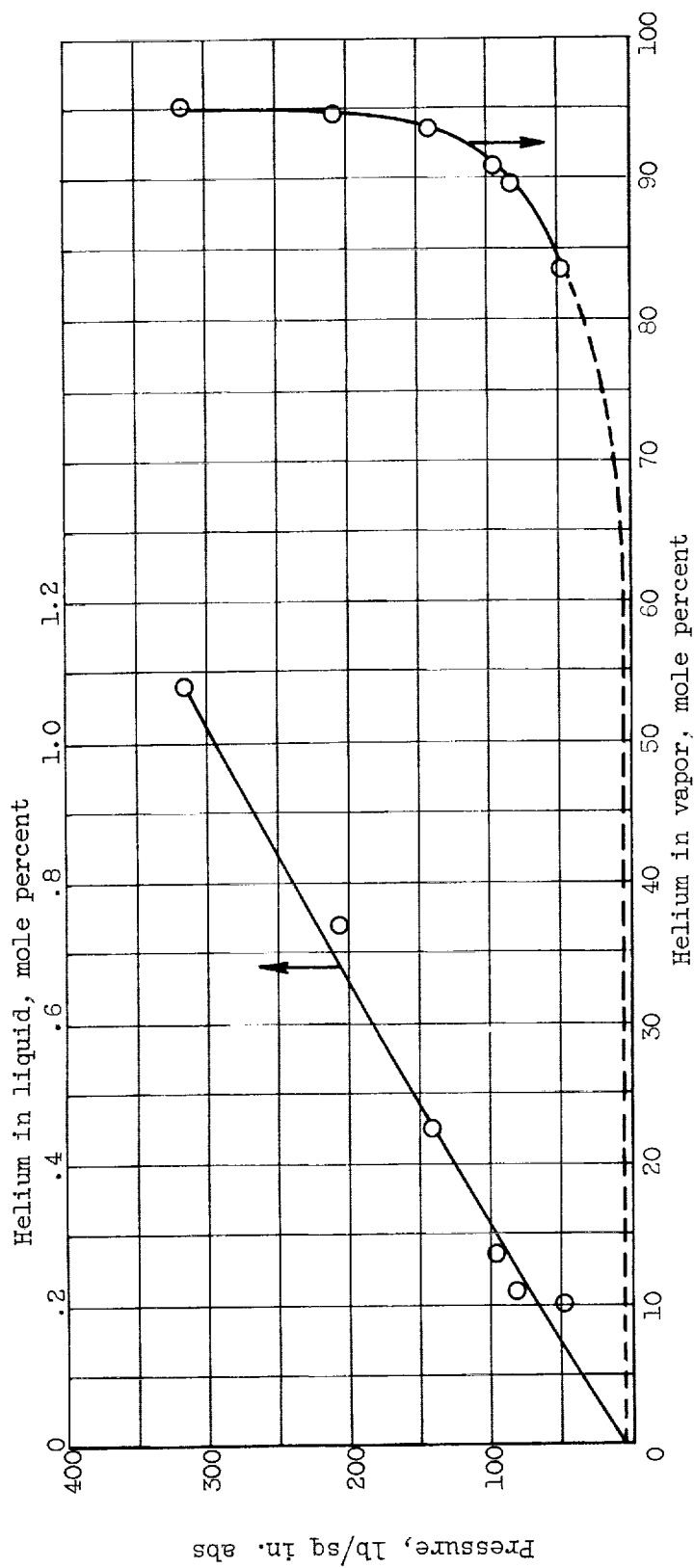
(c) Temperature, 21.8° K; vapor pressure of liquid hydrogen, 1.48 atmosphere; fugacity of liquid hydrogen, 1.33 atmosphere

Pressure, lb/sq in. abs	Helium in liquid, mole percent (a)	Helium in vapor, mole percent (a)	Fugacity of helium, atm (a)	Hydrogen in liquid, mole percent	Gas-phase fugacity of hydrogen, atm (a)	Liquid-phase fugacity of hydrogen, atm (b)	Gas-phase fugacity of hydrogen, atm (c)
43	0.18	60.8	1.78	100	0.957	1.360	1.32
82	.29	73.4	4.08	99.82	1.06	1.415	.985
157	1.36	83.3	8.90	99.71	.940	1.530	1.165
227	1.49	87.9	13.6	98.64	.720	1.650	1.250
306	2.44	88.2	18.35	98.51	.679	1.78	1.205
386	3.06	88.3	23.3	97.56	.586	1.93	1.37
				96.94			1.475

^aRef. 1.

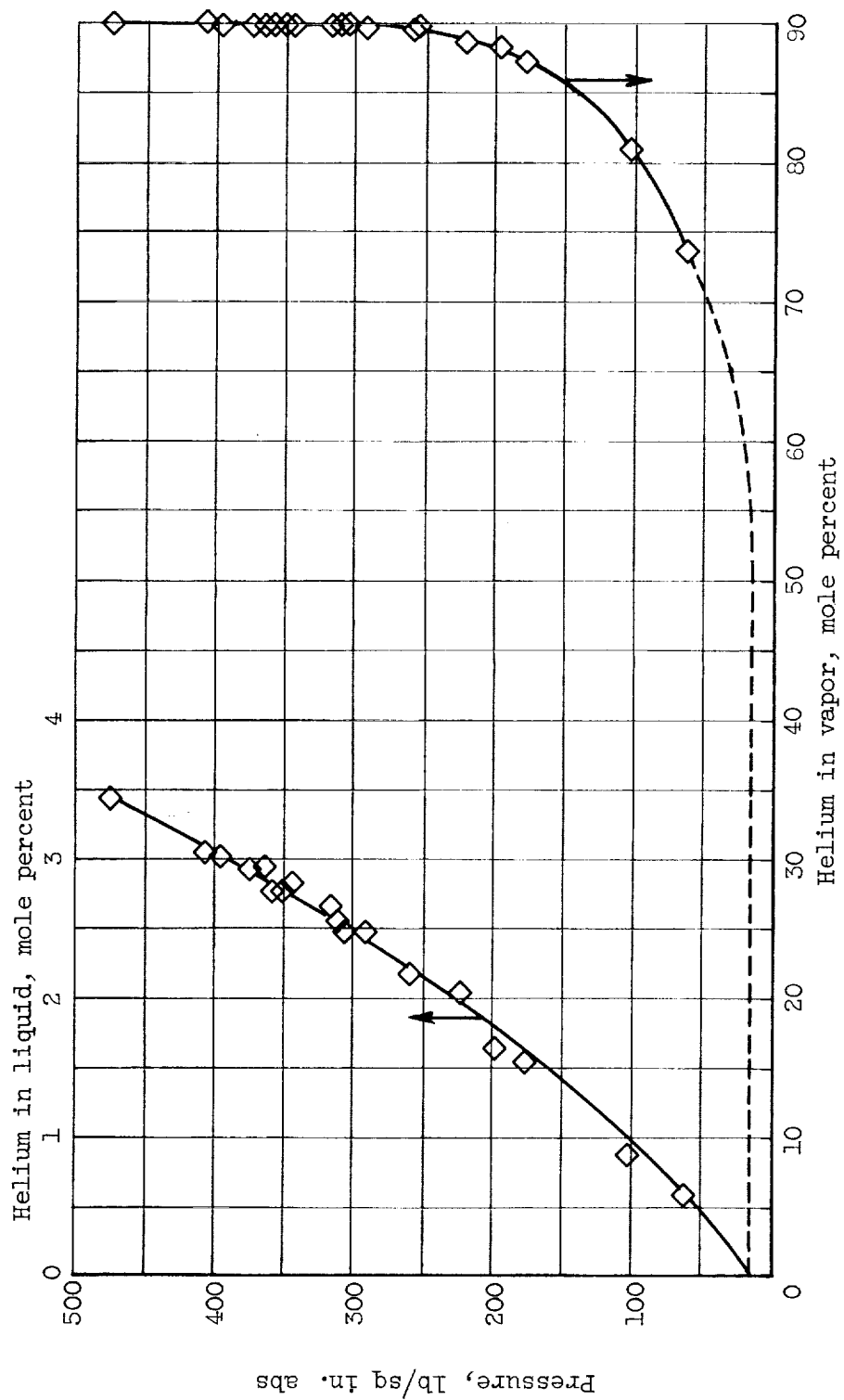
^bEq. (5).

^cCalculated from eq. (13) with virial coefficients of ref. 2.



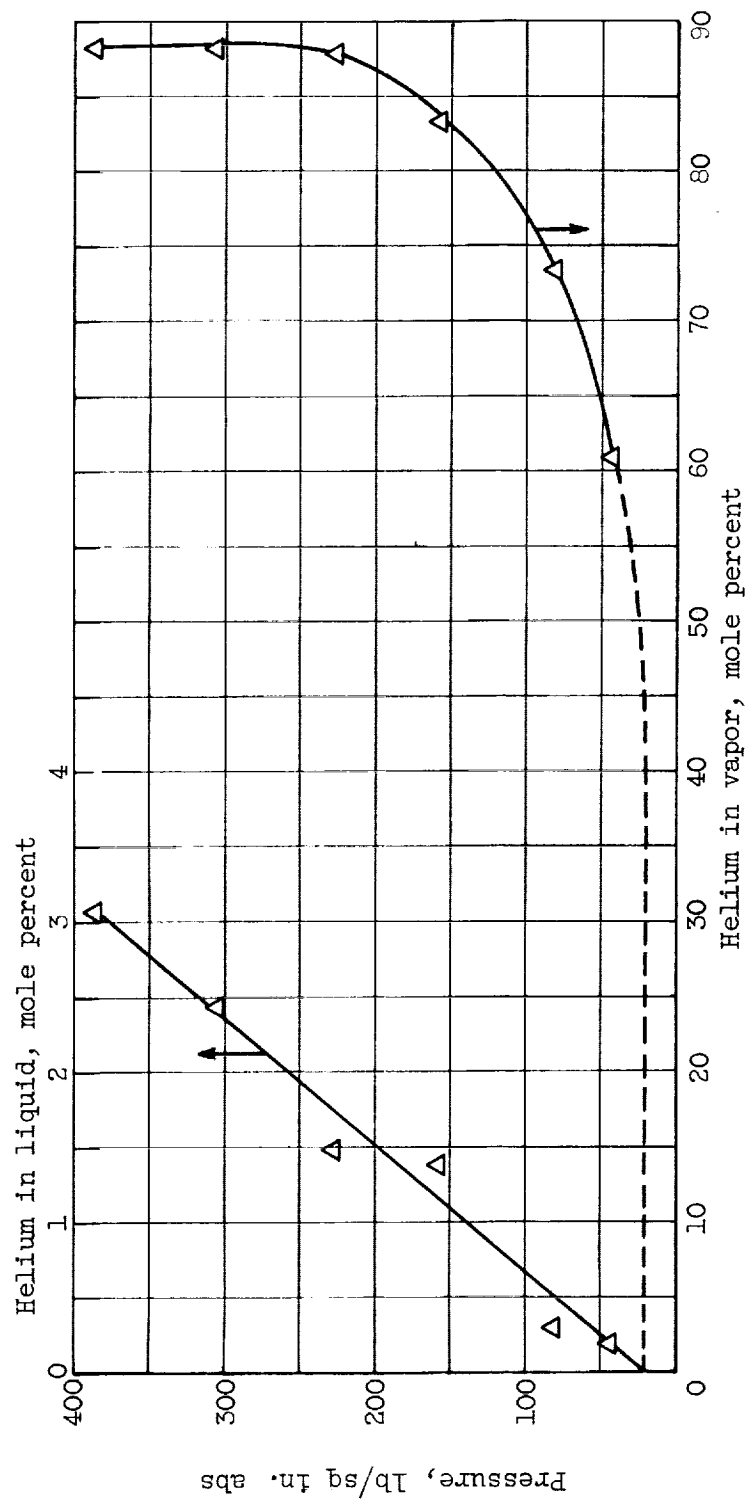
(a) Temperature, 17.4° K.

Figure 1. - Hydrogen-helium vapor-liquid equilibrium data (from ref. 1).



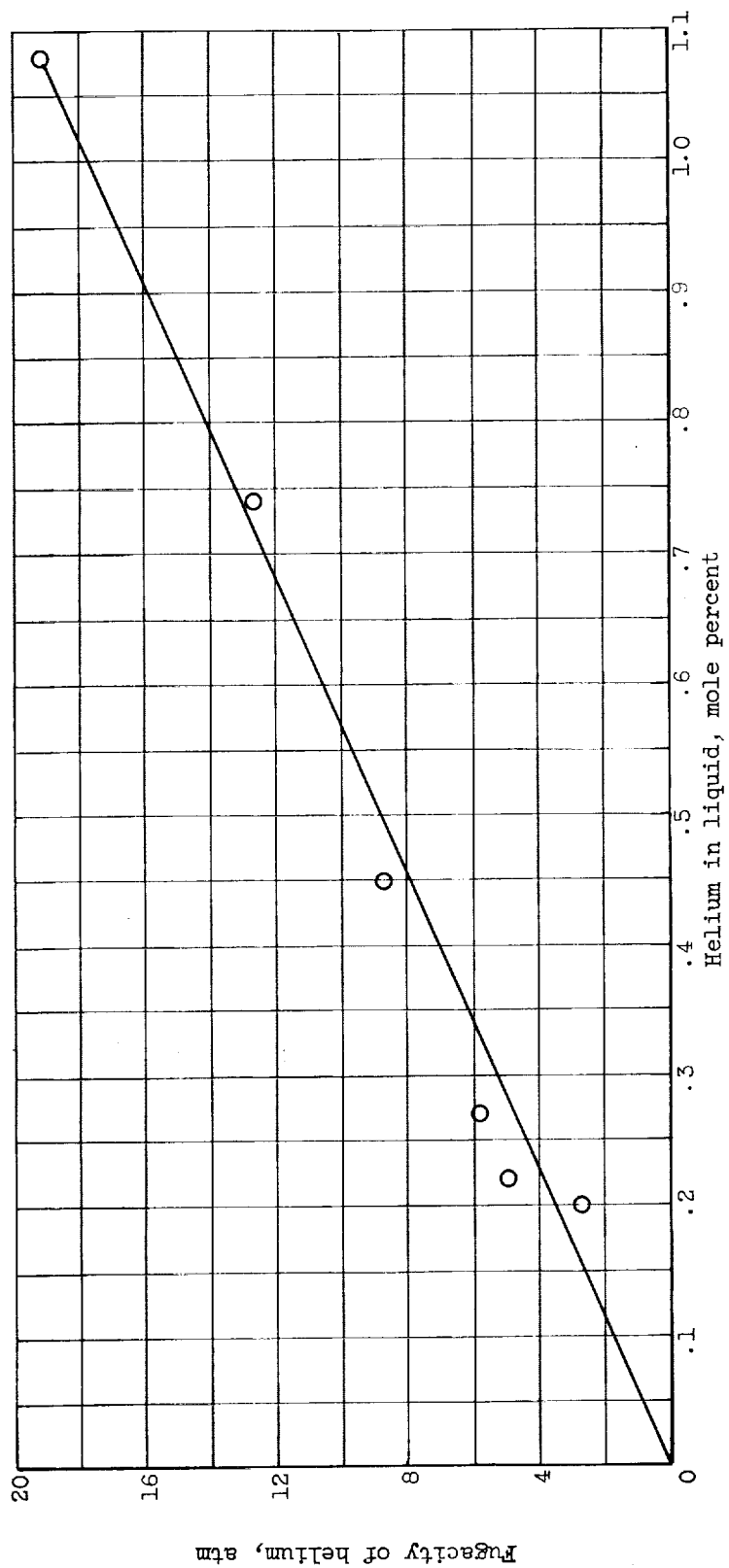
(b) Temperature, 20.39° K.

Figure 1. - Continued. Hydrogen-helium vapor-liquid equilibrium data (from ref. 1).



(c) Temperature, 21.8° K.

Figure 1. - Concluded. Hydrogen-helium vapor-liquid equilibrium data (from ref. 1).



(a) Temperature, 17.4° K.

Figure 2. - Fugacity of helium as function of mole percent of helium. (Data from ref. 1.)

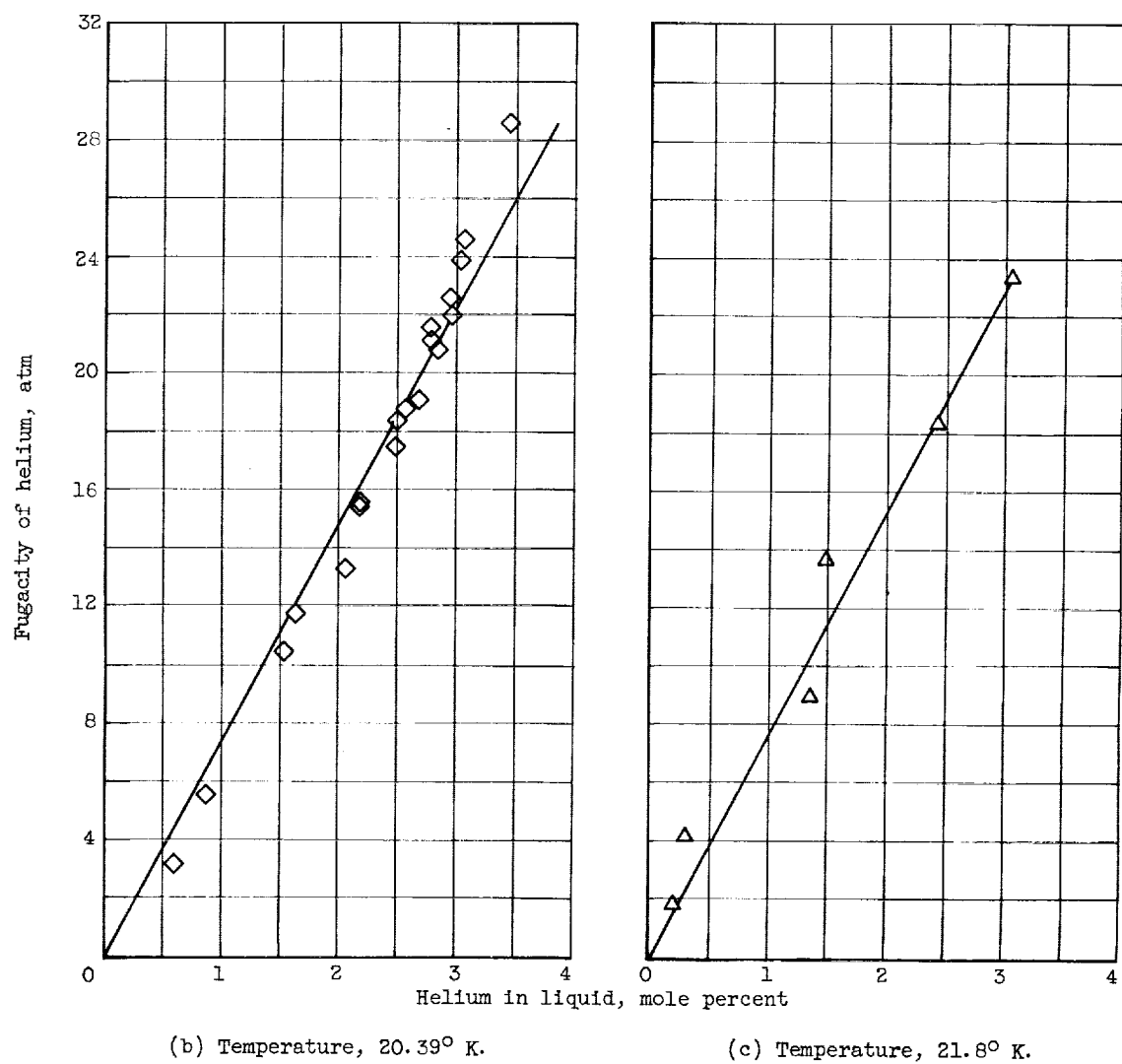
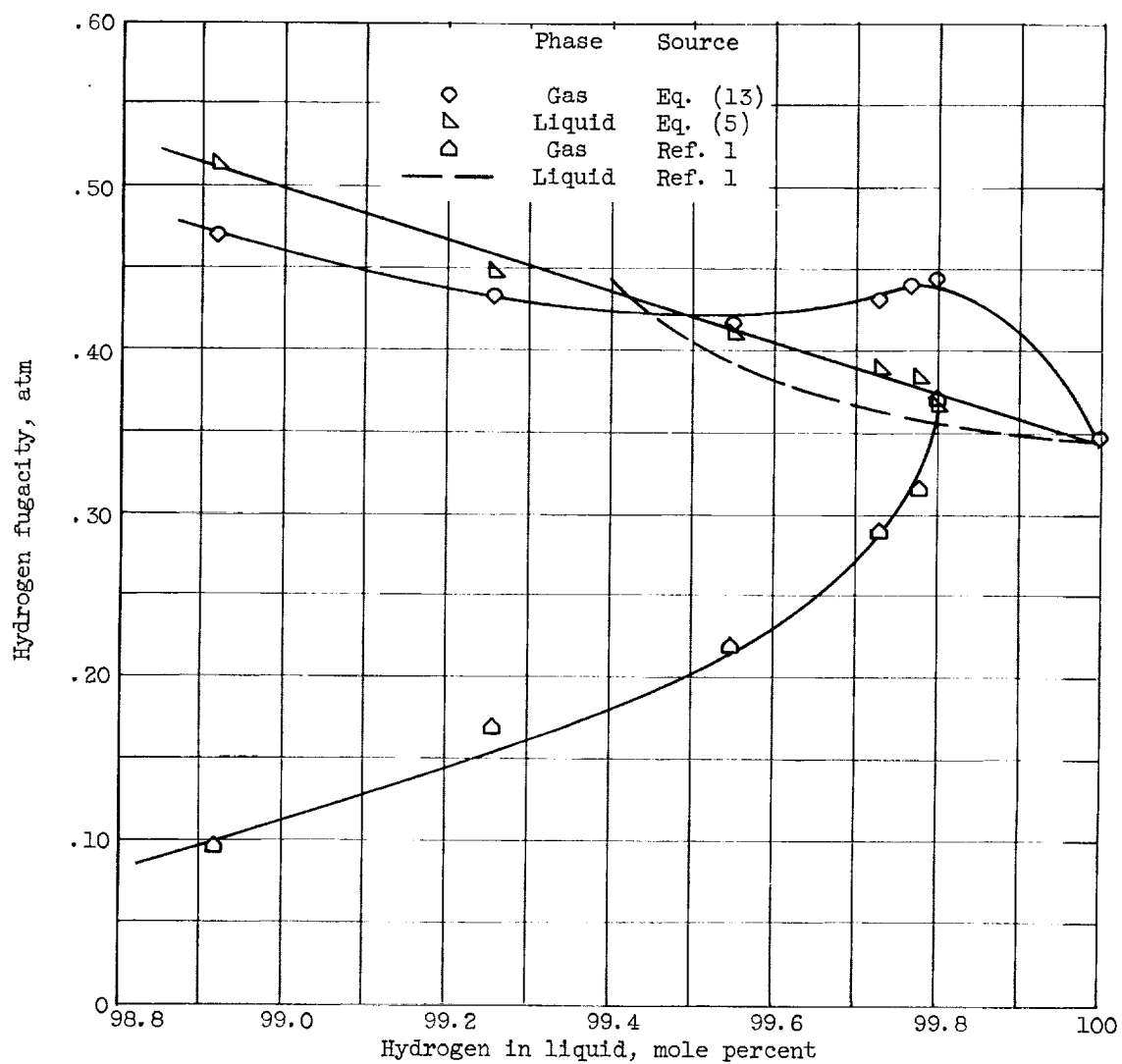
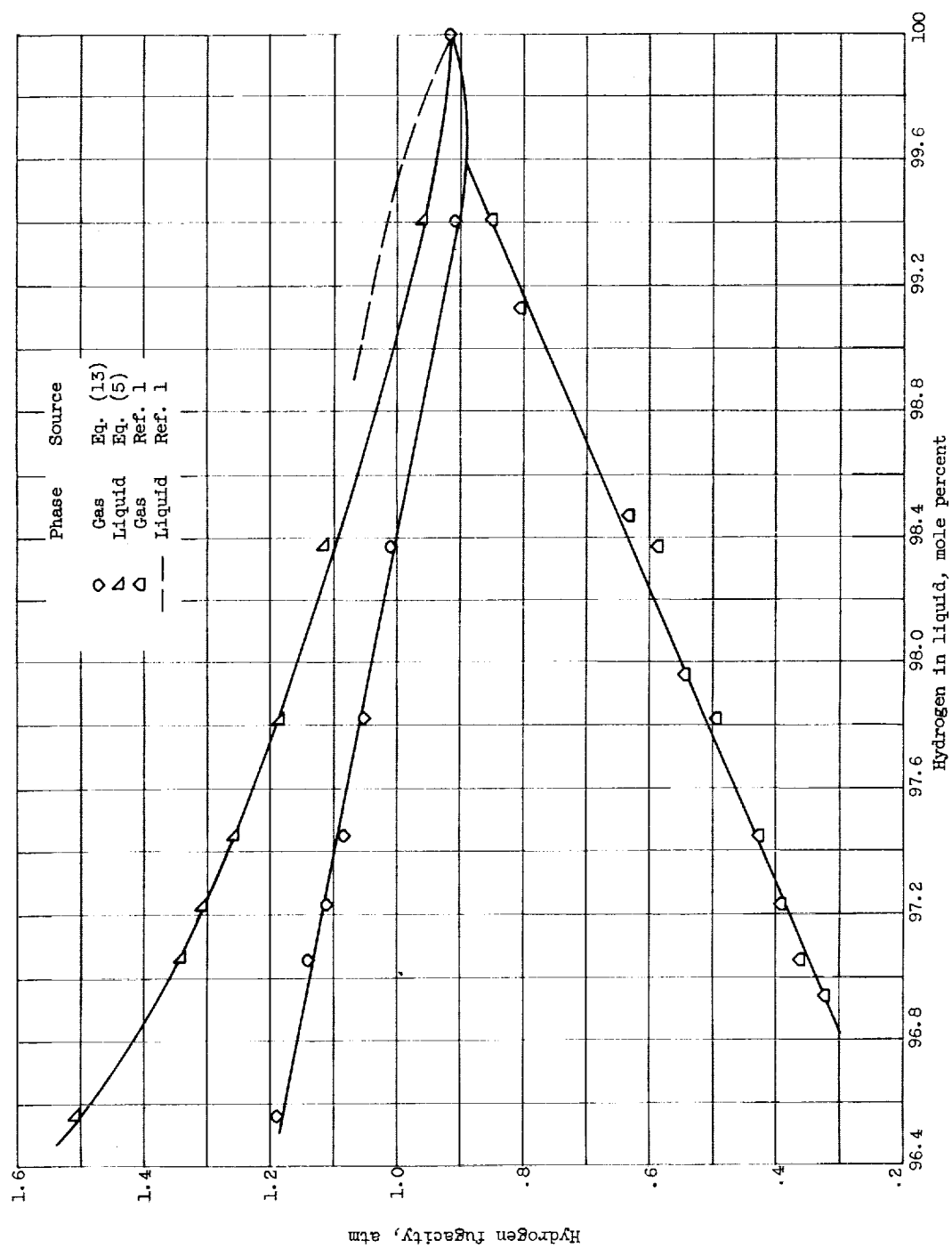


Figure 2. - Concluded. Fugacity of helium as function of mole percent of helium.
(Data from ref. 1.)



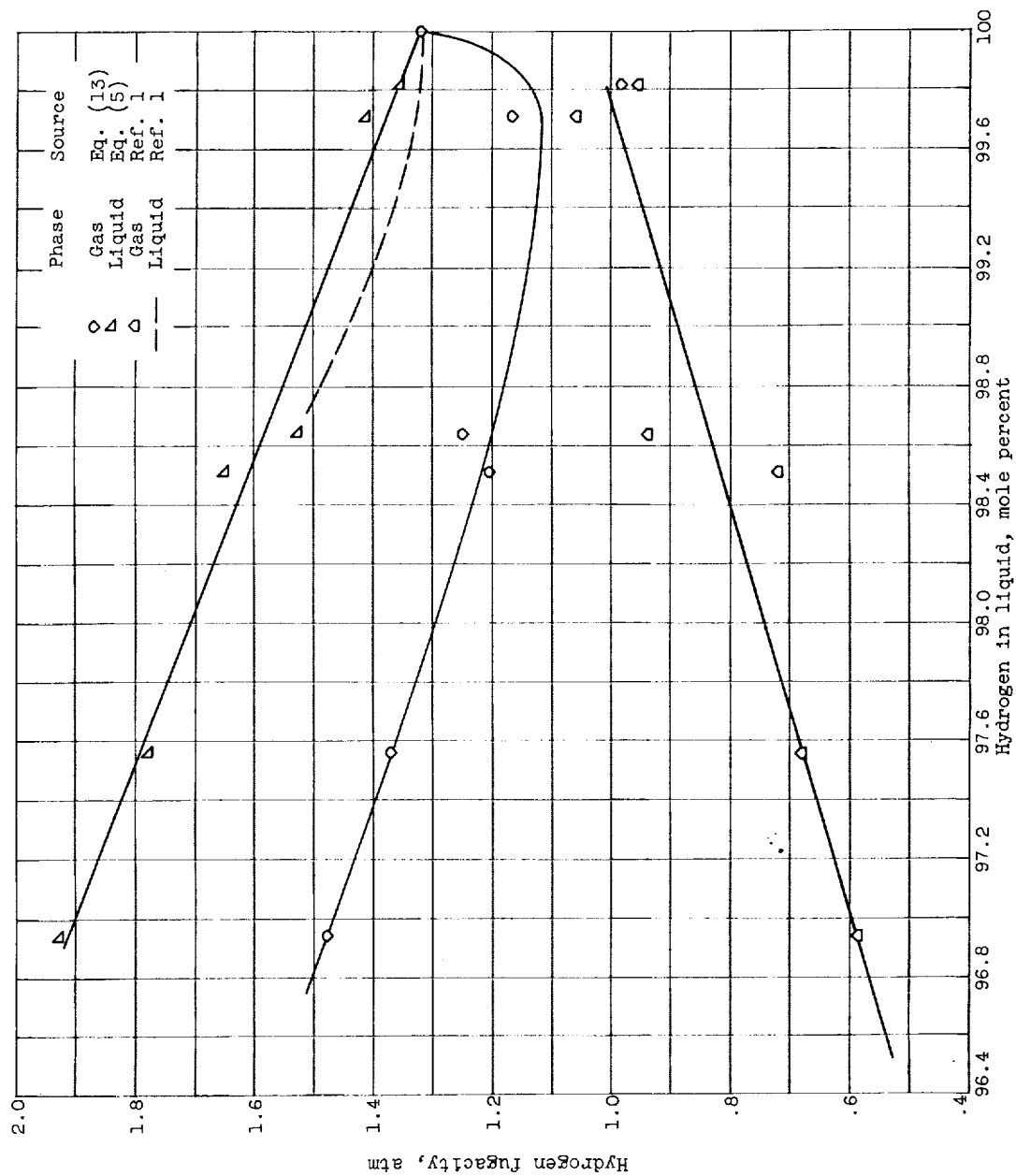
(a) Temperature, 17.4° K.

Figure 3. - Fugacity of hydrogen in hydrogen-helium system.



(b) Temperature, 20.39° K.

Figure 3. - Continued. Fugacity of hydrogen in hydrogen-helium system.



(c) Temperature, 21.8° K.

Figure 3. - Concluded. Fugacity of hydrogen in hydrogen-helium system.

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